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Title: METHODS AND COMPOSITIONS FOR PREPARING SILICA AEROGELS

## In the Claims

- 1. (Withdrawn) A method to prepare a porous cross-linked metal oxide or silicon oxide based aerogel material, the method comprising:
- (a) first, forming a metal oxide or silicon oxide based sol-gel material to provide a preformed metal oxide or silicon oxide based sol-gel material; then,
- (b) contacting the preformed metal oxide or silicon oxide based sol-gel material with a cross-linking agent, the cross-linking agent comprising an organic compound, to provide a cross-linked metal oxide or silicon oxide based sol-gel material comprising organic cross-links; and then,
- (c) drying the cross-linked metal oxide or silicon oxide based sol-gel material to form the porous cross-linked metal oxide or silicon oxide based aerogel material.
- 2. (Withdrawn) The method of claim 1, wherein the cross-linking agent is an isocyanate.
- 3. (Withdrawn) The method of claim 1, wherein the cross-linking agent comprises a diisocyanate, an acid anhydride, an acylchloride, a bis(acid anhydride), or a bis(acylchloride).
- 4. (Withdrawn) The method of claim 1, wherein the cross-linking agent comprises an attached group.
- 5. (Withdrawn) The method of claim 4, wherein the attached group is an absorbant, a catalyst, a fluorophore, a biomolecule, a redox active label, or a reactive group.
- 6. (Cancelled)

- 7. (Withdrawn) The method of claim 1 wherein the cross-linked metal oxide or silicon oxide based aerogel material is a cross-linked silica-based aerogel material.
- 8. (Withdrawn) The method of claim 1, wherein the sol-gel material is substantially filled with a solvent.
- 9. (Withdrawn) The method of claim 8, wherein the solvent is selected from the group consisting of water, ketones, alcohols, esters, carbonates, lactones, hydrocarbons, and mixtures thereof.
- 10. (Withdrawn) The method of claim 9, wherein the alcohol is methanol or ethanol.
- 11. (Withdrawn) The method of claim 9, wherein the ketone is acetone.
- 12. (Withdrawn) The method of claim 9, wherein the solvent is propylene carbonate, ethyl acetate, or butyrolactone.
- 13. (Withdrawn) The method of claim 1, wherein the metal oxide or silicon oxide based solgel material is a silica-based solgel material.
- 14. (Withdrawn) The method of claim 13, wherein the silica-based sol-gel material is prepared from silicon alkoxides via an acid or a base-catalyzed route.
- 15. (Withdrawn) The method of claim 1, wherein the cross-linking agent is in a solvent.
- 16. (Withdrawn) The method of claim 15, wherein the solvent is selected from the group consisting of water, alcohols, ketones, esters, carbonates, lactones, and mixtures thereof.
- 17. (Withdrawn) The method of claim 16, wherein the alcohol is methanol or ethanol.

- 18. (Withdrawn) The method of claim 2, wherein the isocyanate is a monoisocyanate, a diisocyanate, a triisocyanate, a tetraisocyanate, a polyisocyanate, an oligoisocyanate, or a combination thereof.
- 19. (Withdrawn) The method of claim 2, wherein the isocyanate is hexamethylene diisocyanate, poly(hexamethylene diisocyanate), toluene diisocyanate, diphenylmethane diisocyanate, an aliphatic polyisocyanate, triphenylmethyl triisocyanate, or a mixture thereof.
- 20. (Withdrawn) The method of claim 1, wherein the cross-linked metal oxide or silicon oxide based sol-gel material is dried supercritically.
- 21. (Withdrawn) The method of claim 20, wherein the cross-linked metal oxide or silicon oxide based sol-gel material is dried with liquid carbon dioxide.
- 22. (Withdrawn) The method of claim 1, wherein the drying is conducted at ambient pressure.
- 23. (Previously Presented) A porous cross-linked metal oxide or silicon oxide based aerogel material produced by:
- (a) first, forming a metal oxide or silicon oxide based sol-gel material to provide a preformed metal oxide or silicon oxide based sol-gel material; then,
- (b) contacting the preformed metal oxide or silicon oxide based sol-gel material with a cross-linking agent, the cross-linking agent comprising an organic compound, to provide a cross-linked metal oxide or silicon oxide based sol-gel material comprising organic cross-links; and then,
- (c) drying the cross-linked metal oxide or silicon oxide based sol-gel material to form the porous cross-linked metal oxide or silicon oxide based aerogel material.

## 24. (Cancelled).

- (Previously Presented) A cross-linked metal oxide or silicon oxide based sol-gel 25. material, wherein a metal oxide or silicon oxide based sol-gel material is formed to provide a preformed metal oxide or silicon oxide based sol-gel material, then a cross-linking agent comprising an organic compound is deposited on surfaces of the preformed metal oxide or silicon oxide based sol-gel material so as to form a cross-linked metal oxide or silicon oxide based sol-gel material comprising organic cross-links.
- (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel 26. material of claim 25, wherein the cross-linking agent comprises at least about 2% by weight of the cross-linked metal oxide or silicon oxide based sol-gel material.
- (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel 27. material of claim 25, wherein the cross-linking agent comprises at least about 5% by weight of the cross-linked metal oxide or silicon oxide based sol-gel material.
- (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel 28. material of claim 25, wherein the cross-linking agent comprises at least about 10% by weight of the cross-linked metal oxide or silicon oxide based sol-gel material.
- (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel 29. material of claim 25, wherein the cross-linking agent comprises at least about 30% by weight of the cross-linked metal oxide or silicon oxide based sol-gel material.

- 30. (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel material of claim 25, wherein the cross-linking agent comprises at least about 50% by weight of the cross-linked metal oxide or silicon oxide based sol-gel material.
- 31. (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel material of claim 25, wherein the cross-linking agent comprises at least about 80% by weight of the cross-linked metal oxide or silicon oxide based sol-gel material.
- 32. 35. (Cancelled)
- 36. (Currently Amended) The sol-gel material of claim 25, wherein the <u>preformed metal</u> oxide or silicon oxide based sol-gel material is silica based.
- 37. (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel material of claim 25, wherein the cross-linking agent is a diisocyanate, a triisocyanate, a polyisocyanate, or a mixture thereof.
- 38. (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel material of claim 25, wherein the cross-linking agent is hexamethylene diisocyanate, poly(hexamethylene diisocyanate), toluene diisocyanate, diphenylmethane diisocyanate, an aliphatic polyisocyanate, triphenylmethyl triisocyanate, or a mixture thereof.
- 39. (Previously Presented) The cross-linked metal oxide or silicon oxide based sol-gel material of claim 25, wherein the cross-linked metal oxide or silicon oxide based sol-gel material is more resistant to rupture under load than the preformed metal oxide or silicon oxide based sol-gel material prior to cross-linking with the cross-linking agent.
- 40. (Previously Presented) A cross-linked metal oxide or silicon oxide based aerogel material formed by drying of the cross-linked metal oxide or silicon oxide based sol-gel material

of claim 25, the drying being carried out using solvent exchange or supercritical drying, or both, such that the aerogel material does not collapse when in contact with a liquid that comprises water, an alcohol, an ether, a hydrocarbon, an ester, a ketone, a carboxylic acid, a phosphoric acid, or a liquefied gas.

- (Previously Presented) The cross-linked metal oxide or silicon oxide based aerogel 41. material of claim 40, wherein the liquefied gas is nitrogen, argon, helium, hydrogen, or oxygen.
- 42. (Previously Presented) The cross-linked metal oxide or silicon oxide based aerogel material of claim 40, wherein the hydrocarbon is kerosene, gasoline, jet fuel, or rocket fuel.
- (Withdrawn) A method to link an attached group to a cross-linked metal oxide or silicon 43. oxide based sol-gel material comprising the method of claim 1 wherein the cross-linking agent comprising an organic compound that includes the attached group.
- 44. (Withdrawn) A capacitor comprising the material of claim 23.
- 45. (Withdrawn) A dielectric comprising the material of claim 23.
- 46. (Withdrawn) An electrical circuit comprising the material of claim 23.
- 47. (Previously Presented) A thermal insulating material comprising the sol-gel material of claim 25.
- (Original) A tile, door, panel, shingle, shutter, beam, cooler, article of clothing, shoe, or 48. boot comprising the thermal insulating material according to claim 47.
- (Previously Presented) A structural material comprising the material of claim 25. 49.

- (Withdrawn) A method to dry the cross-linked metal oxide or silicon oxide based sol-gel 50.
- material of claim 1 comprising: a) washing the cross-linked metal oxide or silicon oxide based sol-gel material, wherein the cross-linked metal oxide or silicon oxide based sol-gel material is filled with a solvent, with a

new solvent to exchange the solvent of the cross-linked metal oxide or silicon oxide based sol-

gel material with the new solvent; and

- b) drying the solvent-exchanged cross-linked metal oxide or silicon oxide based sol-gel material under non-supercritical conditions.
- (Withdrawn) The method of claim 50, wherein the drying is conducted at ambient 51. pressure.
- (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature 52. that is at least about 4°C.
- (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature 53. that is at least about 20°C.
- (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature 54. that is at least about 40°C.
- (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature 55. that is at least about 60°C.
- (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature 56. that is at least about 80°C.
- (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature 57. that is at least about 100°C.

- 58. (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature that is at least about 200°C.
- 59. (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature that is about or less than 300°C.
- (Withdrawn) The method of claim 50, wherein the drying is conducted at a temperature 60. of about 40°C.
- (Withdrawn) The method of claim 50, wherein the drying is by freeze-drying. 61.
- (Withdrawn) The method of claim 50, wherein the new solvent is an organic solvent. 62.
- 63. (Withdrawn) The method of claim 62, wherein the organic solvent comprises a (C<sub>1</sub>-C<sub>20</sub>)alcohol, a (C<sub>1</sub>-C<sub>20</sub>)alkane, a (C<sub>3</sub>-C<sub>20</sub>)cycloalkane, a (C<sub>2</sub>-C<sub>20</sub>)alkene, a (C<sub>3</sub>-C<sub>20</sub>)cycloalkene, a (C<sub>2</sub>-C<sub>20</sub>)alkyne, an aryl, a (C<sub>1</sub>-C<sub>20</sub>) alkane substituted aryl, a (C<sub>2</sub>-C<sub>20</sub>)alkene substituted aryl, or a  $(C_2-C_{20})$ alkyne substituted aryl.
- (Withdrawn) The method of claim 62, wherein the organic solvent is a  $(C_5-C_{10})$  alkane. 64.
- 65. (Cancelled).
- (Withdrawn) The method of claim 1 wherein chemical functionality of surfaces of said 66. metal oxide or silicon oxide based sol-gel material acts as a template for reaction with the crosslinking agent.
- 67. (Withdrawn) The method of claim 1 wherein surfaces of the metal oxide or silicon oxide based sol-gel material act as a template for the cross-linking agent.

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- 68. (Cancelled).
- 69. (Previously Presented) The metal oxide or silicon oxide based sol-gel material of claim 25 wherein a chemical functionality of the surfaces of said metal oxide or silicon oxide based sol-gel material acts as a template for reaction with, accumulation of, or both, the cross-linking agent.
- 70. (Previously Presented) The metal oxide or silicon oxide based sol-gel material of claim 25 wherein the surfaces of the metal oxide or silicon oxide based sol-gel material act as a template for the cross-linking agent.
- 71. (Previously Presented) The sol-gel material of claim 25 comprising an attached group wherein the cross-linking agent comprising an organic compound includes the attached group.